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A Consideration of the Relationship among the three Types of Lattice in Calcic Plagioclases

By

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Abstract

The transitional and body-centred anorthite lattices can be derived from the primitive anorthite lattice by means of mathematical treatment, presuming the existence of stacking mistakes occurring in a direction along the *b*-axis. As a matter of course, it comes to a conclusion that calcic plagioclases constitute a domain structure, which is in accordance with the current view.

Introduction

During a decade since 1950, researches on the structure of plagioclases had been carried out intensely by using an X-ray, urged by the observations made by TAYLOR *et al.* (1934) and CHAO and TAYLOR (1940). TAYLOR *et al.* (1934) observed that the length of the *c*-axis of anorthite is twice the length of that of albite. CHAO and TAYLOR (1940) observed that intermediate plagioclases gave peculiar reflexions which did not obey to Bragg's law. In the present paper, however, the author will be concerned about the plagioclases in a calcic region.

Observations made by COLE *et al.* (1951), GAY (1953), GAY and TAYLOR (1953), LAVES and GOLDSMITH (1954a, 1954b, 1954c), GAY (1954), GOLDSMITH and LAVES (1955), LAVES and GOLDSMITH (1955), GOLDSMITH and LAVES (1956) and BOWN and GAY (1958) can be summarized as described below.

There are three types of lattice, viz., primitive, transitional and body-centred lattices in plagioclases whose *An* contents are in a range from 70 – 75% to 100%. In the primitive lattice, four kinds of reflexion appear:
(a)-reflexions ($h+k = 2n$, $l = 2n$), very strong and usually sharp,
(b)-reflexions ($h + k = 2n + 1$, $l = 2n + 1$), medium and usually sharp,
(c)-reflexions ($h + k = 2n$, $l = 2n + 1$), rather strong and sometimes diffuse,
(d)-reflexions ($h + k = 2n + 1$, $l = 2n$), very weak, few in number and sometimes diffuse. In the transitonal lattice, three kinds of reflexion appear:
(a)-reflexions ($h + k = 2n$, $l = 2n$), very strong and usually sharp,
(b)-reflexions ($h + k = 2n + 1$, $l = 2n + 1$), medium,
(c)-reflexions ($h + k = 2n$, $l = 2n + 1$), rather weak and diffuse elongated parallel

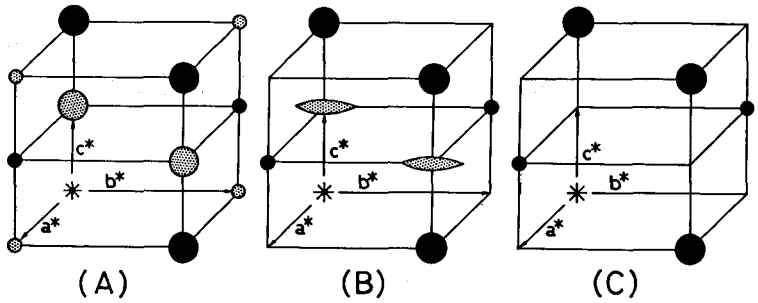

Fig. 1. Schematic representation of three types of lattice recognized in calcic plagioclases. (A): primitive lattice, (B): transitional lattice, (C): body-centred lattice. Large solid circles denote (a)-reflexions, small solid circles (b)-reflexions, large dotted circles and dotted lens-like bodies (c)-reflexions, small dotted circles (d)-reflexions.

to b^*. In the body-centred lattice, two kinds of reflexion appear:
(a)-reflexions ($h + k = 2n$, $l = 2n$), very strong and usually sharp,
(b)-reflexions ($h + k = 2n + 1$, $l = 2n + 1$), medium. These lattices are pictured visually as shown in Fig. 1.

In natural plagioclases, the primitive lattice is characteristic in those whose *An* contents are more than about 95%, the transitional lattice is characteristic in those whose *An* contents are in a range from 80 – 85% to 90 – 95%, the body-centred lattice is characteristic in those whose *An* contents are in a range from 70 – 75% to 80 – 85%. The ranges are, however, not necessarily certain. (More recently, MOSSMAN (1970) suggested that the boundary between the primitive and transitional lattice might lie at 90.5 – 93.0% *An*.)

When plagioclases with the primitive lattice are heated at above 1100 °C, the (c)-reflexions become diffuse, the (d)-reflexions apparently disappear probably becoming diffuse, too. The diffuse character of the (c)-reflexions increases as the temperature is raised until near the melting point, elongating parallel to b^*. Namely, a transformation from primitive lattice to transitional lattice takes lace by heat-treatment. The transformation is relatively rapid and reversible. Decrease in an *An* content promotes the transformation. Observations of the effects by heat-treatment on plagioclases with the other lattices are rather scanty. GAY (1954) reported that a plagioclase with the transitional lattice transformed to that with the body-centred lattice at about 1400 °C and a plagioclase with the body-centred lattice transformed to that with a high albite lattice at 1350 °C. In any case, these transformations take place gradually, not abruptly. GAY (1962) gave a schematic diagram which represents the transformation by heat-treatment as shown in Fig. 2.

BOWN and GAY (1958), CHANDRASEKHAR *et al.* (1961) and GAY (1962) are doubtful as to whether the body-centred plagioclase is a stable equilibrium form at an ordinary temperature.

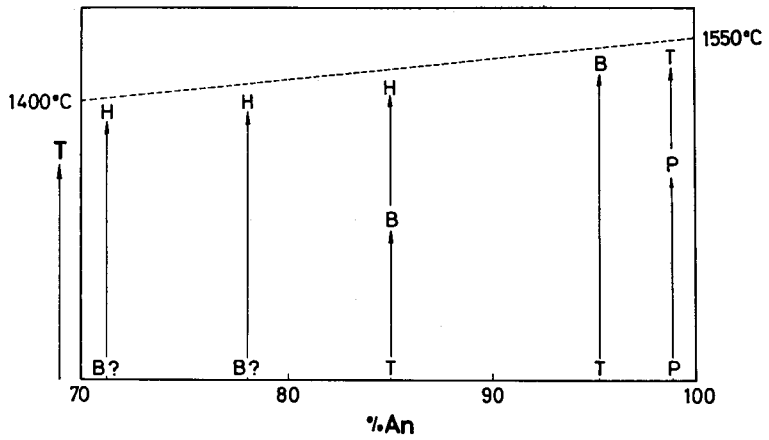

Fig. 2. Schematic representation of sub-solidus relationships from 70% to 100% *An*. Dotted line denotes the solidus. P: primitive lattice, T: transitional lattice, B: body-centred lattice, H: high albite lattice. (After GAY, somewhat modified.)

GAY and TAYLOR (1953) considered that the transformation was due to the *Si-Al* arrangement, in the primitive lattice the arrangement being ordered, in the transitional lattice it being partially ordered and in the body-centred lattice it being disordered. While, GOLDSMITH and LAVES (1955) and LAVES and GOLDSMITH (1955) reported that the *Si-Al* arrangement was ordered in both the primitive and transitional lattices. GOLDSMITH and LAVES (1956) considered that a formation of the body-centred lattice was due to out-of-step *Ca* atom domains.

During the next decade since 1960, crystal structure analyses and interpretations had been tried on the three modifications by several researchers. Crystal structure of a plagioclase with the body-centred lattice, whose *An* content is 72 – 80% was determined by SÖRUM (1953), by CHANDRASEKHAR *et al.* (1961) and by FLEET *et al.* (1966). Crystal structure of a plagioclase with the primitive lattice, whose *An* content is 95 – 100% was determined by KEMPSTER *et al.* (1962), by MEGAW *et al.* (1962) and by WAINWRIGHT and STARKEY (1971). Crystal strucutre of a plagioclase with the transitional lattice, whose *An* content is 99% (This specimen is a volcanic ejecta.) was determined by RIBBE and MEGAW (1962).

In the primitive lattice, *Si* and *Al* tetrahedra are alternate, viz., the *Si-Al* arrangement is ordered, the same is the case in the transitional lattice, too. In both the lattice, no differences in atomic coordinates can be recognized. In the body-centred lattice, *Si* and *Al* atoms possess definite positions respectively in the lattice, that is, the *Si-Al* arrangement is ordered, although atomic coordinates are slightly different from those of the other two. In addition to these facts, it should be necessary to notice that the three lattices are almost the same in the cell-dimensions and

the axial angles. Slight differences in the cell-dimensions and the axial angles are attributed to the difference of *An* content. Interpretations about the three modifications made by several researchers will be referred later on in connection with considerations.

Recently, the author has found that the transitional and body-centred lattices can be derived from the primitive lattice mathematically. In the derivation, the author presumed that there exist stacking mistakes occurring in a direction along the *b*-axis within a crystal. As already described above, in the primitive lattice the (c)- and (d)- reflexions are sometimes diffuse, but in a primitive lattice of a pure anorthite which crystallized at an ideal condition, the both reflexions might not be diffuse. In the derivation the author will assume that in an ideal primitive lattice the reflexions are all sharp.

Mathematical consideration

Cut a unit-cell into two halves, cutting plane being parallel to (010). The left side sub-cell will be denoted by X and the right side one by Y. Exchange the sub-cells so as to arrange as YX. The original cell will be denoted by P and the new cell by Q'. Let the structure factor of P be F. Then, it can be proved that the structure factor of Q' is $F \cdot \exp 2\pi i(k/2)$. Suppose that at the beginning sub-cells stack in a manner as XYXYXY....., but that there is a chance α of stacking in a manner as YXYXYX....., until there is another mistake which restores the stacking as XYXYXY..... It will be assumed that such a manner of stacking mistakes takes place in a direction along the *b*-axis as shown in Fig. 3. Clearly, such a manner of stacking mistakes can be represented as PPP.....Q'Q'Q'.....PPP.....

Transfer Q' with a shift of $1/2\,\vec{a} + 1/2\,\vec{c}$. The Q' transferred to the new position

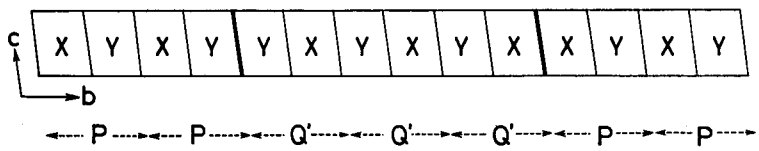

Fig. 3. Showing the stacking of sub-cells in which mistakes have taken place along the *b*-axis direction.

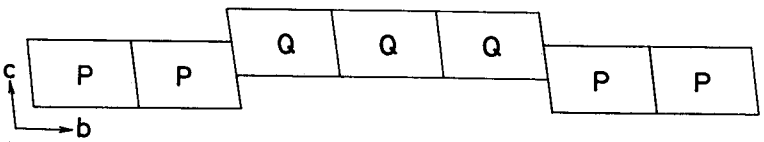

Fig. 4. Showing the stacking of unit-cells in which mistakes have taken place along the *b*-axis direction. Q's are shifted by an amount of $1/2\vec{a}+1/2\vec{c}$.

will be denoted by Q. Then, the structure factor of Q is $F\cdot\exp 2\pi i(k/2)\cdot\exp 2\pi i(h/2+l/2) = F\cdot\exp 2\pi i(h/2+k/2+l/2)$.

Assume that unit-cells stack in a manner as PPPP.....QQQQ.....PPPP..... in a direction along the *b*-axis as shown in Fig. 4.

[*Remark*] Consider a unit-cell whose origin is transferred to the body center of an original one. Denote the latter by A and the former by B. Suppose that at the beginning stacking is in a manner as AAAA....., but that there is a chance α of stacking in a manner as BBBB....., until there is another mistake which restores the stacking as AAAA..... Assume that such a manner of stacking mistakes takes place in a direction along the *b*-axis. The result obtainable is the same as that obtained through the process described above.

Let P_m be the probability of the mth added cell being P, and Q_m the probability of its being Q. Then,

$$P_m+Q_m=1 \tag{1}$$

$$P_{m+1}=\alpha+(1-2\alpha)P_m. \tag{2}$$

P_{m+1} means the probability of the $(m+1)$th added cell being P. Since starting cell is P, then, from the equations (1) and (2) we get

$$P_m=\frac{1}{2}\{1+(1-2\alpha)^m\} \tag{3}$$

$$Q_m=\frac{1}{2}\{1-(1-2\alpha)^m\}. \tag{4}$$

As the structure factor of P is F and that of Q is $F\cdot\exp 2\pi i(h/2+k/2+l/2)$ the structure factor of the mth added cell, F_m, is

$$F_m=P_mF+Q_mF\cdot\exp 2\pi i(h/2+k/2+l/2). \tag{5}$$

Let the real part, A_0, and the imaginary part, B_0, of the structure factor of the starting cell be as

$$\left.\begin{aligned} A_0&=F\\ B_0&=0\end{aligned}\right\} \tag{6}$$

respectively. From the equation (5) it is clear that the real part, A_m, and the imaginary part, B_m, of the structure factor of the mth added cell are

$$\left.\begin{aligned} A_m&=P_mF+Q_mF\cos\pi(h+k+l)\\ B_m&=Q_mF\sin\pi(h+k+l)\end{aligned}\right\} \tag{7}$$

respectively. Let A_0, A_1, A_2,....., B_0, B_1, B_2,..... be, respectively, the real parts and imaginary parts of the structure factors of the unit-cells arranged succeedingly along the *b*-axis direction. The total intensity of a reflexion as a function of v can be approximated, according to WILSON (1962), as follows, in so far as the chance of a mistake is large compared with $1/N$, but small compared with $1/2$, where N is the number of unit-cells.

$$I(v)=2N\int_0^\infty(J_m\cos 2\pi vm-K_m\sin 2\pi vm)dm, \tag{8}$$

where, v is a small fraction of the distance between two reciprocal points parallel to b^*, J_m is the mean value of the form $A_nA_{n+m}+B_nB_{n+m}$ and K_m is that of the form $A_nB_{n+m}-A_{n+m}B_n$. It can be proved that the mean value of $A_nA_{n+m}+B_nB_{n+m}$ is reduced to $A_0A_m + B_0B_m$ and that of $A_nB_{n+m}-A_{n+m}B_n$ is to $A_0B_m-A_mB_0$, where, A_0, A_m and B_0, B_m are the same things as said above respectively. In the present case, therefore, we get the following equations using the relations of (6) and (7),

$$\left.\begin{aligned} J_m&=A_0A_m+B_0B_m=F^2\{P_m+Q_m\cos\pi(h+k+l)\}\\ K_m&=A_0B_m-A_mB_0=F^2\{Q_m\sin\pi(h+k+l)\}. \end{aligned}\right\} \quad (9)$$

Replacing P_m and Q_m in the above equations by the values shown in (3) and (4),

$$\left.\begin{aligned} J_m&=F^2\left[\frac{1}{2}\{1+(1-2\alpha)^m\}\right.\\ &\qquad\left.+\frac{1}{2}\{1-(1-2\alpha)^m\}\cos\pi(h+k+l)\right]\\ K_m&=F^2\left[\frac{1}{2}\{1-(1-2\alpha)^m\}\sin\pi(h+k+l)\right]. \end{aligned}\right\} \quad (10)$$

Hence, when $h + k + l = 2n$,

$$\left.\begin{aligned} J_m&=F^2\left[\frac{1}{2}\{1+(1-2\alpha)^m\}+\frac{1}{2}\{1-(1-2\alpha)^m\}\right]\\ &=F^2\\ K_m&=0. \end{aligned}\right\} \quad (11)$$

The results indicate that the reflexions with $h + k + l = 2n$ (that is, $h + k = 2n$ and $l = 2n$, or $h + k = 2n + 1$ and $l = 2n + 1$, the former corresponds to the (a)-reflexions and the latter to the (b)-reflexions) do not suffer any effect even if there exist stacking mistakes under consideration.

On the other hand, when $h + k + l = 2n + 1$,

$$\left.\begin{aligned} J_m&=F^2\left[\frac{1}{2}\{1+(1-2\alpha)^m\}-\frac{1}{2}\{1-(1-2\alpha)^m\}\right]\\ &=F^2(1-2\alpha)^m\\ K_m&=0. \end{aligned}\right\} \quad (12)$$

In such a case equation (8) becomes to

$$I(v)=2N\int_0^\infty J_m\cos2\pi vmdm \quad (13)$$

Replacing J_m in the above equation by the value shown in (12), the equation (13) becomes to

$$I(v)=2NF^2\int_0^\infty(1-2\alpha)^m\cos2\pi vmdm. \quad (14)$$

The above equation can be solved as follows, when α is in a range as $0<\alpha<1/2$,

$$I(v) = 2NF^2 \frac{-\log(1-2\alpha)}{(2\pi v)^2 + \{\log(1-2\alpha)\}^2}. \tag{15}$$

$$\left.\begin{aligned} \text{Accordingly, } I &= \int_{-\infty}^{\infty} I(v)dv = 2NF^2 \{-\log(1-2\alpha)\} \frac{1}{-2\log(1-2\alpha)} \\ &= NF^2, \end{aligned}\right\} \tag{16}$$

$$\text{and} \quad I_0 = \frac{2NF^2}{-\log(1-2\alpha)}. \tag{17}$$

Then, integral breadth: $\sigma = I/I_0 = -1/2 \log (1 - 2\alpha)$.

Hence, when $\alpha = 0$, $\sigma = 0$,
when $\alpha \fallingdotseq 0$, $\sigma = \alpha$,
when $\alpha = 1/2$, $\sigma = \infty$.

Because of the results obtained above, when the probability of α, the chance of a mistake, is equal to zero, the reflexions with $h + k + l = 2n + 1$ (that is, $h + k = 2n$ and $l = 2n + 1$, or $h + k = 2n + 1$ and $l = 2n$, the former corresponds to the (c)-reflexions and the latter to the (d)-reflexions) are sharp; when the probability of α is nearly equal to zero, but not equal to zero, they become diffuse along b^* direction forming streaks, the greater the probability of α, the more they become diffuse; and when the probability of α is equal to 1/2, they fade into the background, that is, practically they disappear.

In summary; when stacking mistakes occur in a direction along the b-axis, the shift being $1/2\vec{a} + 1/2\vec{b} + 1/2\vec{c}$ (speaking briefly, see *remark* described above), (i) in so far as the probability of α, the chance of a mistake, is nearly equal to zero, the (c)-reflexions become diffuse in streaks along b^* direction, the degree of diffuseness depending on the probability of α, the (d)-reflexions disappear fading into the background on account of the fact that they are originally very weak, as the result cell becomes transitional as shown in Fig. 1-(B), (ii) if the probability of α is equal to 1/2 (in practice, α need not necessarily to be equal to 1/2, a little greater value than that of (i) is sufficient), both the (c)- and (d)-reflexions disappear fading into the background, in such a case cell becomes body-centred as shown in Fig. 1-(C).

Considerations

The author considers, looking into the structure of anorthite, that a stacking mistake which the author has pictured may be plausible. It should be noticed that stacking a Q′-cell on a P-cell in the direction along the b-axis, shifting by an amount of $1/2\vec{a} + 1/2\vec{c}$ is equivalent to stacking a B-cell on an A-cell in the direction along the b-axis. (Connotations of A, B, P and Q′ have already been described.) Therefore, a stacking mistake under consideration can be treated as one-dimensional·

The author has derived the transitional and body-centred lattices from the primitive lattice by a mathematical treatment, presuming the existence of stacking mistakes occurring in a direction along the *b*-axis. As a matter of course, it can be said that calcic plagioclases are built up of domains, the size of the domain being dependent upon the probability of α, the chance of a mistake.

GAY and TAYLOR (1953) considered that the transitional and body-centred lattices arise from a disorder in the *Si-Al* arrangement. However, the results of crystal structure analyses made by SÖRUM (1953), CHANDRASEKHAR *et al.* (1961), KEMPSTER *et al.* (1962), MEGAW *et al.* (1962), RIBBE and MEGAW (1962), FLEET *et al.* (1966) and WAINWRIGHT and STARKEY (1971) show that the *Si-Al* arrangement is ordered in the transitional and body-centred lattices as well as in the primitive lattice. GAY and TAYLOR (1953) inferred that the change of lattice type by the heat treatment had a bearing on *Ca* atom arrangement. GOLDSMITH and LAVES (1956) considered that the body-centred lattice arose from out-of-step *Ca* atom domains. These researchers, however, gave nothing from which they arrived to such considerations. MEGAW (1959) emphasized the importance of an effect of stacking fault on the diffraction and MEGAW (1960) tried to consider the effect by mathematical treatment. The treatment, however, aimed to interpret the so-called non-Bragg reflexions which appear in intermediate plagioclases. MEGAW (1961) also emphasized that a puckering of the ordered *Si-Al* framework was a principal cause of the transformation from the primitive to the body-centred lattice, in the paper published in the next year MEGAW (1962) stated that the formation of the three modifications was due to a domain structure arising from stacking faults which took place at the boundaries of sub-cells. RIBBE and COLVILLE (1968) interpreted the anisotropic diffuseness of the (c)-reflexions of the transitional lattice as a result of out-of-step domains. LAVES *et al.* (1970) and CZANK *et al.* (1970) proposed that anorthite was composed of two kinds of domain, one is made up of A-cells, the other is of B-cells (after their notation). The latter is that whose origin is chosen at the body-centre of the former. They, however, did not prove the proposition neither by an experimental observation nor by a theoretical treatment. MÜLLER *et al.* (1972) were successful to observe domain structure in anorthite from various origin by using a high voltage electron microscope. Size of the domains was varied by the chemical composition and the thermal and mechanical history.

The cause by which calcic plagioclases are apt to form domain structure is an important problem to be solved hereafter. MEGAW *et al.* (1962) suggested that the formation of domains originated in a site mistake of *Ca* atoms. The transformation observed by BROWN *et al.* (1963) and FOIT and PEACOR (1967) in anorthite when it was heated at 350 °C, or when it was quenched after heating at above 1100 °C was interpreted by them as a result of decreasing anisotropy of the bonding of *Ca* atoms

to the neighbouring oxygen atoms as the temperature rises. SMITH and RIBBE (1969) are trying to interpret the formation of the three modifications from the standpoint of kinetics.

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